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The influence of the interlayer on the magnetic and structural properties of three-layer systems

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Abstract. This article is dedicated to the influence analysis of the thickness and composition of nonmagnetic intermediate layers, t_{NM}, on the magnetic and structural properties of Co/Mo/Co, Co/Si/Co, Co/Bi/Co and Fe_1/PDP/Fe_2 thin-film systems, as well as comparing the mechanisms of exchange interaction between ferromagnetic, FM, layers through NM spacers. The thicknesses of Co layers are equal to 5.0 nm and the thickness of Fe layers is varied from 14.0 to 50.0 nm. The thickness of NM layers, depending on its composition, is changed from 0.2 to 50.0 nm. It is found that the hysteresis loops for some samples measured in a magnetic field applied parallel to the easy magnetization axis have a rectangular form and for others – more complex two-step ones. The saturation field, H_S, of Co/Mo/Co , Co/Si/Co and Co/Bi/Co samples oscillates in magnitude. These data are explained by the presence of exchange coupling between the Co layers through a NM spacer. Fe_1/PDP/Fe_2 samples have a two-step hysteresis loops, which at $t_{PDP} \leq 10$ nm are explained by the exchange coupling between the Fe layers through the PDP layer, and at $t_{PDP} > 10$ nm - by the magnetostatic interaction between the Fe layers due to the difference in their thickness.

1. Introduction

Physical properties of thin magnetic films of 3d-transition metals and magnetic multilayers materials are the most important area of the physics of magnetic phenomena and applied magnetism. This is due to the rapid development of fundamental knowledge of such phenomena as quantum size effects [1, 2], giant magnetoresistance [3, 4] and oscillating exchange coupling between ferromagnetic layers (Fe, Co) through nonmagnetic (for example, Cu, Cr, Ag and Au) spacers [4, 5], as well as due to the wide practical application of these materials in spintronics and micro- and nanoelectronics devices. A significant improvement in the methods of obtaining and studying of thin-film systems with well-controlled thicknesses of layers of the nanometer range contributed to these researches [6].

Despite the existing large volume of experimental data, the study of the magnetic properties of thin-film magnetic structures still attracts the attention of researchers from both a scientific and practical points of view. At the same time, it should be noted that early investigations of systems were mainly devoted to the study of multilayer structures but the presence of a large number of layers in previously studied systems, as a rule, complicated the interpretation of experimental data.

This problem was solved by a detailed study of three-layer samples consisting of two ferromagnetic, FM, layers (Co or Fe) and various nonmagnetic, NM, intermediate layers, in particular, metallic (Mo), semiconductor (Si), semimetallic (Bi) and polymeric (poly (diphenylenephthalide), PDP) and comparing their magnetic properties.

This article is dedicated to the influence analysis of the thickness and composition of foregoing nonmagnetic intermediate layers, t_{NM}, on the magnetic and structural properties of three-layer thin-film systems, as well as comparing the mechanisms of exchange interaction between FM layers through nonmagnetic layers.

2. Sample and Experimental method

The Co/Mo/Co, Co/Si/Co and Co/Bi/Co samples were grown by magnetron sputtering at room temperature using Co, Mo, Si and Bi targets and corning 2845 glass substrates. In the Co/Bi/Co samples the Ta seed layers of 5 nm thick were deposited on the glass substrates. The background pressure in the vacuum chamber was $4 \cdot 10^{-7}$ mbar and the argon pressure was as high as $3.8 \cdot 10^{-3}$ mbar. The constant magnetic field was applied parallel to the substrate, H_{SUB} = 250 Oe, in order to form a uniaxial magnetic anisotropy.

The Fe_1/PDP/Fe_2 systems were obtained by using the spin-coating method. To produce PDP layers of various thicknesses the solutions of polymer in cyclohexanone with different concentrations were used. It should be noted that polymer solutions with a concentration of 1–10% allow to obtain polymer films with thickness from 5.0 nm to 1.3 mm. The thickness of Co, Fe and nonmagnetic intermediate (Mo, Si, Bi, PDP) layers of studied samples are presented in table 1.

Table 1. Parameters of the samples.

Samples	t_{FM}, nm (each layer)	t_{NM}, nm
Co/Mo/Co	5.0	0.5 – 4.0
Co/Si/Co	5.0	0.2 – 3.2
Co/Bi/Co	5.0	0.2 – 50.0
Fe_1/PDP/Fe_2	14.0 – 50.0	10.0 – 35.0

The microstructure of the samples based on Co was studied by X-ray diffraction (XRD) using CuKα radiation. The surface morphology of all samples was investigated by atomic force microscopy (AFM). The magnetic characteristics of the studied samples were measured employing magneto-optical magnetometer and vibration sample magnetometer with the sensitivity up to 10^{-5} G•cm^3. The magneto-optical measurements were carried out via the transverse Kerr effect (TKE), proportional to the magnetization component parallel to the sample surface and perpendicular to the plane of incidence of light. The hysteresis loops of the studied samples were measured at two directions of the external magnetic field. In one case, the H orientation was parallel to the magnetic field direction which had been applied during the deposition process (direction D1), and in the other – perpendicular to D1 (direction D2).

The domain structure (DS) of the samples was investigated by the magneto-optical Kerr magnetometer (MOKM) made on the basis of a Carl Zeiss polarization microscope. MOKM allows to measure the hysteresis loops and to visualize changes of the DS during the magnetization reversal at the same time. MOKM measurements were carried out in the geometry of meridional Kerr effect, proportional to the magnetization component which is parallel to the sample surface and the plane of light incidence. All measurements in this work were performed at the room temperature.

3. Results and discussion

The results of XRD measurements showed that Co layers in all studied samples have a nanocrystalline structure. According to AFM investigations, the average surface roughness R_a of the studied samples does not exceed 0.6 nm, and the magnitude of R_a does not depend on t_{NM}. These data testify the high quality of sample surfaces. Moreover, this experimental fact shows that the surface roughness should not practically influence the magneto-optical and magnetic properties of the samples under study.

It was found that hysteresis loops measured in the magnetic field, applied parallel to the D1 and D2 directions differ from each other. This fact indicates the formation of induced magnetic anisotropy with the easy magnetization axis parallel to the direction of magnetic field, applied during the deposition process. According to the commonly accepted notions, the main mechanism underlying the appearance of induced magnetic anisotropy is the pair ordering of atoms [7].

Figure 1 shows the most typical hysteresis loops observed at a *H* orientation parallel to D1. The results of magnetic measurements showed that the shape of the hysteresis loops of three-layer samples, in a magnetic field, applied parallel to the direction D1, depends on t_{NM} (figure 1).

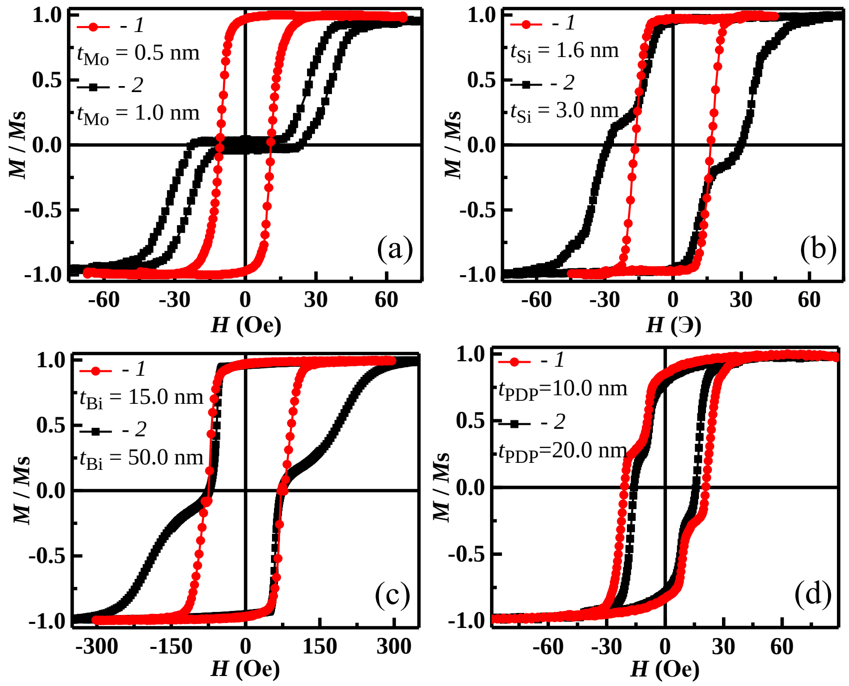

Figure 1. Typical hysteresis loops measured at *H* parallel to D1 direction for samples: (a) Co/Mo/Co; (b) Co/Si/Co; (c) Co/Bi/Co; (d) Fe_1/PDP/Fe_2 (t_{Fe1} =16 nm, t_{Fe2} = 15 nm).

In particular, for some values of t_{NM}, the rectangular hysteresis loops are observed, and for others t_{NM} more complex two-step ones. According to the reported experimental and calculated data [8, 9], almost rectangular and two-step hysteresis loops are observed for parallel and antiparallel orientations of magnetization components in Co layers, respectively. This fact indicates the presence of the ferromagnetic (FM) and antiferromagnetic (AFM) exchange between the magnetic layers through nonmagnetic interlaer.

The magnetization reversal of the samples with the rectangular hysteresis loops proceeds via irreversible growth of the remagnetization nuclei. Abrupt change in optical contrast during the remagnetization process from $-H$ to $+H$ ($|H| > H_S$), observed by using MOKM, confirms this fact (figure 2). The measurements of hysteresis loops allowed to obtain the dependencies $H_S(t_{NM})$ for the samples under study (see figure 3).

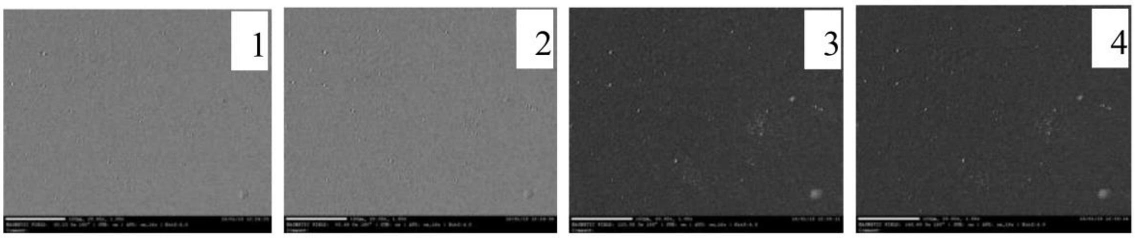

Figure 2. Illustration of abrupt change of optical contrast during the remagnetization process of the sample from $-H$ to $+H$ ($|H| > H_S$), observed by using MOKM.

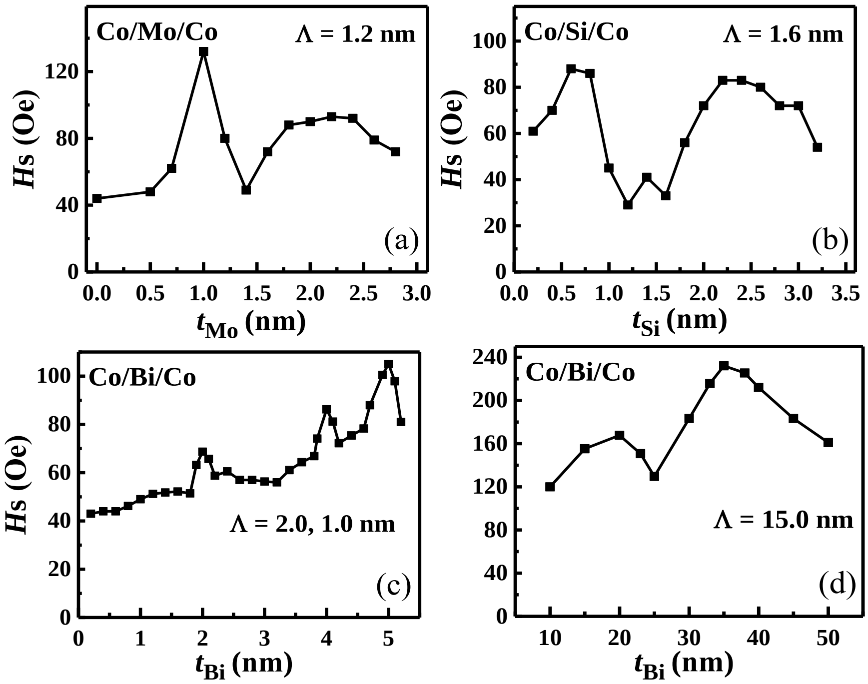

Figure 3. Dependencies of the saturation field H_S on t_{NM}, observed for Co/Mo/Co, Co/Si/Co and Co/Bi/Co samples, at H parallel to D1.

One can see that H_S oscillates in magnitude with changing t_{NM}. According to existing data, such behavior of $H_S(t_{NM})$ indicates the presence of ferromagnetic (FM) and antiferromagnetic (AFM) exchange coupling between Co layers through the nonmagnetic (metallic Mo, semiconductor Si, semimetallic Bi) interlayers.

A detailed analysis of the results is given below.

The dependence of $H_S(t_{Mo})$ of Co/Mo/Co samples were explained by the exchange coupling between the Co layers through the Mo nonmagnetic metallic interlayer and its oscillatory behavior as a function of t_{Mo} (figure 3(a)). The maximum values of H_S of Co/Mo/Co samples are observed at t_{Mo} = 1.0 and 2.2 nm. It was found that the oscillation period Λ of H_S, estimated from the difference of the thicknesses of the nearest Mo layers, at which antiferromagnetic (AFM) exchange is observed, of the order of 1.2 nm. It should be noted that it has been experimentally discovered in [10] that Λ reduces with decreasing thickness of the Co layer.

When discussing these data, the following information should be taken into attention. It is known that the oscillation period Λ of the exchange coupling between magnetic layers through a nonmagnetic interlayer by means of RKKY interaction should be of the order of π/k_F: $\Lambda \approx \pi/k_F$, where k_F is the

Fermi wave vector [11]. For most metals, π/k_F of the order of 0.3–0.4 nm, which is smaller than the found value of Λ for Co/Mo/Co samples.

The presence of a longer period of H_S oscillations in thin-film systems with metal nonmagnetic interlayers has been explained by the influence of quantum size effects manifesting in a change of the electronic structure of the ultrathin magnetic layer (the appearance of so-called Quantum Well States, QWSs) compared with bulk samples [12, 13]. In this case, the oscillation period Λ should be equal to $\pi/[k_{BZ} - k_F]$, where k_{BZ} is the value of the vector to the Brillouin zone boundary in the direction perpendicular to the film surface. Taking into account the above relation, it has been found in [5, 14] that the value of Λ of the order of 1.0-2.0 nm.

The saturation field of the samples with an AF exchange, H_S^{AF}, is significantly larger than H_S of the samples with FM exchange (see figure 3(a)) that is caused by the additional expenditure of energy to overcome the AFM exchange between the magnetic layers. According to the calculations made in [12, 15], the value of this field can calculate by using the relation:

$$H_S^{AF} \approx 4\, J_{AF} / M_S\, t_{FM} \;\Rightarrow\; |J_{AF}| \approx H_S^{AF}\, M_S\, t_{FM} /4, \qquad (1)$$

where J_{AF} is the constant of AFM exchange between the magnetic layers. The calculation of the maximum value of $J^{MAX}{}_{AF}$ by using the formula above showed that for the Co/Mo/Co samples $J^{MAX}{}_{AF} \approx 0.084$ erg / cm^2. This value is almost an order of magnitude smaller than the J_{AF} observed for Co/Ru thin-film systems in [15]. The obtained $J^{MAX}{}_{AF}$ value can be explained by a decrease of the H_S value observed for the studied samples, as compared with previously studied in the works of other authors. In turn, the decrease in H_S can be explained by the nanocrystalline structure of the samples under study, which, according to calculations [16], can reduce H_S on several orders of magnitude.

Finally, according to [12], the exchange between the magnetic layers through a nonmagnetic metal layer is carried out by polarized conduction electrons of the nonmagnetic layer interacting with the magnetic moments of the magnetic layers (indirect exchange of the RKKI type).

In the case of Co/Si/Co samples the maximum values of H_S has been observed at t_{Si} = 0.6 and 2.4 nm (figure 3(b)). The variations of H_S with increasing t_{Si} have been explained with taking into account the structural peculiarities of these systems It is known [17], that at a small thickness of t_{Si}, the interlayer consists of the silicides of cobalt. With increasing t_{Si}, the silicon layer appears that is accompanied by the growth of H_S. Structural changes in the Si layer influence the magnetic-field behavior of the Co/Si/Co samples. At t_{Si} < 1.6 nm Co/Si/Co samples exhibit almost rectangular hysteresis loops (figure 1(b), loop 1). In this case, at small values of H, the stray fieldcreated by cobalt layers, affect the magnetic-field behavior of the Co/Si/Co samples. In particular, they reduce the effective value of the external magnetic field, acting on the sample that causes the H_S increase. However, with increasing t_{Si}, the effect of stray fields decreases that is accompanied by a decrease in H_S.

The dependence of $H_S\,(t_{Si})$ at t_{Si} > 1.6 nm has the following features. The values of H_S grow with increasing t_{Si} up to 2.4 nm and then decreases. The Co/Si/Co samples with $t_{Si} \approx 2.4$ nm exhibit two-step hysteresis loops (figure 1(b), loop 2). This fact indicates that in the above range t_{Si} there is a transition from FM to AFM and back to FM exchange between magnetic layers through the Si interlayer (FM–AFM–FM transition). The oscillation period of H_S, estimated from the distance between the thicknesses of silicon, at which the FM exchange is observed, was about 1.6 nm. This value of Λ is greater than π/k_F.

The above allows us to conclude that the dependence $H_S\,(t_{Si})$ at t_{Si} > 1.6 nm can be explained by a change in the antiferromagnetic exchange constant J_{AF} between cobalt layers with increasing t_{Si} [12, 15]. An estimate of $J^{MAX}{}_{AF}$ value showed that for the Co/Si/Co samples $J^{MAX}{}_{AF} \approx 0.016$ erg / cm^2. This value is almost an order of magnitude smaller than the J_{AF} observed for Fe/Si thin-film systems in [18].

The exchange mechanism between magnetic layers with a semiconductor interlayer differs from the exchange mechanism in samples with a metallic layer. In particular, according to existing data, the possible mechanism of the exchange coupling between the Co layers via the Si interlayer is the tunneling of polarized electrons through localized defects, which are always present in the semiconductor layer.

The $H_S\,(t_{Bi})$ dependence of three-layer Co/Bi/Co samples, had also oscillatory behavior (figures 3(c), 3(d)), which indicated the presence of ferromagnetic and antiferromagnetic exchange between Co layers through a nonmagnetic intermediate layer in these samples. Moreover, it was found that the maximum H_S values increase with increasing t_{Bi}. The results of structural studies of thin-film Co/Bi/Co systems allowed to explain this fact. It was established experimentally that texturing of the samples under study increases with increasing t_{Bi}, which is accompanied by an increase in H_S values [19].

It was found that the peaks of H_S are observed at $t_{Bi} = 2.0$, 4.0, 5.0 (figure 3 (c)), 20.0, 35.0 (figure 3(d)) nm. The distances between peaks, Λ, are equal to 2.0, 1.0 and 15.0 nm. Hysteresis loops of the samples with the above t_{Bi} had a double-step form (figure 1 (c), loop 2), which testified the existence of AFM exchange interaction between ferromagnetic Co layers through the Bi interlayer. The magnitude of $J^{MAX}{}_{AF}$ was estimated for the Co/Bi/Co samples with $t_{Bi} = 2.0$ nm since in this case the influence of the Bi texture on H_S was minimum. It was found that $J^{MAX}{}_{AF} \approx 0.013$ erg/cm^2. This J_{AF} value is almost two orders of magnitude smaller than J_{AF} of thin-film systems with a metal spacer [15]. The small J_{AF} value might have been caused by the low carrier concentration of semimetal Bi. In particular, at $T = 300$ K, the concentration of charge carriers of Bi equal to n = p = $5{\bullet}10^{18}$ cm^{-3}.

The oscillations of $H_S\,(t_{Bi})$ were observed with different periods. Herewith the range of oscillations increased up to the bismuth thickness equal to 50.0 nm which is significantly more than observed in the thin-film systems with metallic interlayers. When discussing the oscillation behavior of $H_S\,(t_{Bi})$ using the exchange interaction mechanism RKKY [11, 20], the peculiarities of bismuth structure must be taken into account. In particular, the electronic properties of Bi are fundamentally different from the electronic properties of metals due to the complex and highly anisotropic Fermi surface. The elongated pockets of holes and electrons with small effective masses lead to a large Fermi wavelength $\lambda_F \approx 40.0$ nm, as opposed to a few Å in most metals. The mean free path of charge carriers in Bi can reach several millimeters at 4.2 K, which is several orders of magnitude larger than that of most metals [21]. The calculated oscillation period of H_S for Co/Bi/Co samples using the ratio $\Lambda \sim \lambda_F/2$ of order 20.0 nm is much longer than the experimentally found value. Moreover, the experimentally found period of H_S oscillations in Co/Bi/Co samples is not constant. The reason of the oscillation period change can be the dependence of the Fermi energy on t_{Bi} [22], which arises from the nonparabolic band structure of bismuth, and also the changes in the band structure of the thin Bi layer as compared with a bulk sample [23, 24].

Finally, appropriate to analyse the results of study of the magnetic-field behavior of the three-layer Fe_1/polymer (polydiphenylenephthalide – PDP)/Fe_2 samples. The hysteresis loops for these sample were measured in a magnetic field, applied in the plane of the samples parallel to their 10-mm length or 7-mm width (indicated also as D1 and D2, respectively). The hysteresis loops, measured along the D1 direction, had also a two-step shape wherein a step size depended on the difference of the thicknesses of Fe layers: the more difference, the more the step on the two-step hysteresis loops. The two-step hysteresis loops testified antiparallel orientation of the magnetization in the Fe layers. It should be noted that the above two-step hysteresis loops were observed in the region of the thickness of the PDP layer from 10.0 to 35.0 nm. Let us dwell on the discussion of the obtained data. It is known that submicron PDP films even without the use of chemical doping methods can exhibit atypical electrophysical properties, manifesting in high metal-like conductivity. It was shown in [25] that in PDP materials, the conductivity under the action of minor external influences can reach values comparable to the conductivity of metals. Moreover, thin-film structures containing a PDP layer can react to changes in the external magnetic field. In particular, when a nonmagnetic PDP layer is deposited on a ferromagnetic substrate, its conductivity increases by several orders of magnitude (up to 12–16) [26]. The foregoing allowed to suggest that the physical reason of the exchange interaction between the magnetic layers through the PDP layer is similar the observed in the thin- film magnetic systems with nonmagnetic metallic layer.

Further the following fact was taken into account. Experimental studies of thin-film systems, presented, for example, in [27], testified to the presence of exchange interaction between magnetic layers through a nonmagnetic metal intermediate layer up to 7.0 nm thick, while photoemission spectroscopy data – up to 10.0 nm [28, 29]. Thus, it can be assumed that the reason of two-step hysteresis loops observed at $t_{PDP} \leq 10.0$ nm is the exchange interaction between the magnetic layers through the PDP layer, which is carried out by polarized conduction electrons of the nonmagnetic layer interacting with the magnetic moments of the magnetic layers [12].

At the same time, taking into account the data of paper [9], the two-step hysteresis loops observed for the Fe_1/PDP/Fe_2 samples with $t_{PDP} > 10.0$ nm can be explained by a difference of the thickness of magnetic layers. Taking into account the data, presented in [30], the reason of above hysteresis loops is the magnetostatic interaction between the magnetic layers through the PDP layer.

4. Conclusions

The influence analysis of the thickness and composition of nonmagnetic intermediate layers, t_{NM}, on the magnetic and structural properties of three-layer Co/Mo/Co, Co/Si/Co, Co/Bi/Co and Fe_1/PDP/Fe_2 thin-film systems has been carried out. The comparison of the mechanisms of exchange interaction between FM layers through nonmagnetic spacer has been performed. These experimental results can be useful at the development of new layered structures for practical applications.

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